SO(4) Theory of Antiferromagnetism and Superconductivity in Bechgaard Salts

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Motivated by recent experiments with Bechgaard salts, we investigate the competition between antiferromagnetism and triplet superconductivity in quasi one-dimensional electron systems. We unify the two orders in an SO(4) symmetric framework, and demonstrate the existence of such symmetry in one-dimensional Luttinger liquids. SO(4) symmetry, which strongly constrains the phase diagram, can explain coexistence regions between antiferromagnetic, superconducting, and normal phases, as observed in $(TMTSF)_2PF_6$. We predict a sharp neutron scattering resonance in superconducting samples.

A common feature of many strongly correlated electron systems is proximity of a superconducting state to some kind of magnetically ordered insulating state. Examples include organic materials[1, 2], heavy fermion superconductors[3, 4], and high T_c cuprates[5]. Several theoretical analyses suggest that a strong repulsion between the two orders plays an important role in determining the phase diagram and low energy properties of these materials[6]. The idea of competing orders was developed into the SO(5) theory of high T_c superconductivity of S.C. Zhang[7]. SO(5) symmetry has also been applied to study competition of ferromagnetism and p-wave superconductivity in Sr_2RuO_4[8], and antiferromagnetism and d-wave superconductivity in κ-BEDT-TTF salts[9].

In this paper we consider the interplay of antiferromagnetism (AF) and triplet superconductivity (TSC) in quasi one-dimensional (Q1D) electron systems. Our study is motivated by Q1D Bechgaard salts $(TMTSF)_2X$. The most well studied material from this family, $(TMTSF)_2PF_6$, is an antiferromagnetic insulator at ambient pressure and a superconductor at high pressures[10, 11, 12, 13]. The symmetry of the superconducting order parameter in $(TMTSF)_2PF_6$ is not yet fully established, but there is strong evidence that electron pairing is spin triplet: the superconducting T_c is strongly suppressed by disorder[14]; critical magnetic field H_{c2} in the interchain direction exceeds the paramagnetic limit[15]; the electron spin susceptibility, obtained from Knight shift measurements, does not decrease below T_c[16]. In another material from this family, $(TMTSF)_2ClO_4$, superconductivity is stable at ambient pressure and also shows signatures of triplet pairing[17, 18, 19, 20]. Insulator to superconductor transition as a function of pressure has also been found in$(TMTSF)_2AsF_6$[21].

The phase diagram of interacting electrons in one dimension was obtained in Ref. 22 using bosonization and renormalization group (RG) analyses. At incommensurate filling, this system has a phase boundary between spin density wave (SDW) and TSC phases when $K_\rho = 1$ and $g_1 > 0$ (K_ρ is the Luttinger parameter in the charge sector, g_1 the backward scattering amplitude). The starting point of our discussion is the observation that, in the absence of umklapp, 1D Luttinger liquids have an "isospin" $SO(4)_{\rm iso}$ symmetry[23] at the boundary between SDW and TSC phases. To define this symmetry, we introduce the charge of left and right moving electrons, $Q_\pm$, and two new operators, $\Theta_\pm$, $(r = \pm)$

$$\begin{aligned} Q_r &= \frac{1}{2}\sum_{ks}\left(a^\dagger_{r,ks}a_{r,ks} - \frac{1}{2}\right) \\ \Theta^\dagger_r &= r\sum_k a^\dagger_{r,k\uparrow}a^\dagger_{r,-k\downarrow}. \end{aligned} \tag{1}$$

Here $a^\dagger_{\pm,ks}$ creates right/left moving electrons of momentum $\pm k_f + k$ and spin s. Combining these according to $J^r_x = (\Theta^\dagger_r + \Theta_r)/2$, $J^r_y = (\Theta^\dagger_r - \Theta_r)/2i$, and $J^r_z = Q_r$, we see that the generators satisfy two independent chiral SO(3) algebras, $\left[J^r_a, J^{r'}_b\right] = i\delta^{r,r'}\epsilon^{abc}J^r_a$. The product of left and right algebras yields the total isospin group $SO(4)_{\rm iso} \approx SO(3)_R \times SO(3)_L$. The Luttinger Hamiltonian at incommensurate filling, $\mathcal{H}$, generically commutes with the charge operators $Q_\pm$. Using a bosonized form of $\mathcal{H}$, it can be shown that at $K_\rho = 1$, the $\Theta_\pm$ operators also commute with $\mathcal{H}$[23, 24]. Thus, at $K_\rho = 1$, $SO(4)_{\rm iso}$ forms an exact symmetry of $\mathcal{H}$. In addition, for spin-symmetric interactions, which describe Bechgaard salts to a very good approximation[24, 25], the system has $SO(3)_{\rm spin}$ symmetry, $[S_\alpha, S_\beta] = i\epsilon^{\alpha\beta\gamma}S_\gamma$, generated by the total spin

$$S_\alpha = \frac{1}{2}\sum_{r,kss'} a^\dagger_{r,ks}\sigma^\alpha_{ss'}a_{r,ks'}. \tag{2}$$

Hence, for $K_\rho = 1$, *i.e.* the line separating SDW and TSC phases, the system has full $SO(3)_{\rm spin}\times SO(4)_{\rm iso}$ symmetry. We emphasize that for Luttinger liquids at incommensurate filling, this symmetry always appears at the SDW/TSC phase boundary and does not require fine-tuning of the parameters.

This symmetry can be used to unify SDW and TSC order parameters. SDW order away from half filling is described by a complex vector order parameter,

$$\Phi_\alpha = \sum_{kss'} a^\dagger_{+,ks}\sigma^\alpha_{ss'}a_{-,ks'}. \tag{3}$$

Q1D band structure restricts the orbital component of triplet superconducting order to be $\vec{\Psi}(\vec{p}) \propto p_x$, with x

the coordinate along the chains. Thus, the TSC order parameter is also a complex vector,

$$\Psi^\dagger_\alpha \;=\; \frac{1}{i}\sum_{kss'} a^\dagger_{+,ks}(\sigma^\alpha\sigma_2)_{ss'}a^\dagger_{-,-ks'} \qquad (4)$$

The four vector order parameters $\mathrm{Re}\vec{\Phi}$, $\mathrm{Im}\vec{\Phi}$, $\mathrm{Re}\vec{\Psi}$, and $\mathrm{Im}\vec{\Psi}$ can be combined into a 4×3 matrix,

$$P_{\bar{a}\alpha} \;=\; \begin{pmatrix} (\mathrm{Re}\vec{\Psi})_x & (\mathrm{Im}\vec{\Psi})_x & (\mathrm{Re}\vec{\Phi})_x & (\mathrm{Im}\vec{\Phi})_x \\ (\mathrm{Re}\vec{\Psi})_y & (\mathrm{Im}\vec{\Psi})_y & (\mathrm{Re}\vec{\Phi})_y & (\mathrm{Im}\vec{\Phi})_y \\ (\mathrm{Re}\vec{\Psi})_z & (\mathrm{Im}\vec{\Psi})_z & (\mathrm{Re}\vec{\Phi})_z & (\mathrm{Im}\vec{\Phi})_z \end{pmatrix} \qquad (5)$$

Each column (row) of $\hat{P}$ transforms independently as a vector under the action of $\mathrm{SO}(3)_{\mathrm{spin}}$ ($\mathrm{SO}(4)_{\mathrm{iso}}$).

The Θ_r operators are reminiscent of Yang's η operator, which generates an SO(4) symmetry for the Hubbard model[26]. Unlike η, whose center of mass momentum is always the commensurate wave vector π, the Θ_r have their momenta at the wave vectors $\pm 2k_f$. This is crucial for defining the symmetry at arbitrary electron density; in contrast, Yang's SO(4) applies only at half-filling.

Real materials are only Q1D and coupling between chains gives rise to finite temperature phase transitions. However, as long as 3D coupling is weaker than the intrachain tunnelling and interactions, the nature of the ordered state is determined by the most divergent susceptibility within individual chains. Hence, for Q1D materials near the SDW/TSC boundary we expect to find K_ρ close to one, and to find approximate $\mathrm{SO}(3)_{\mathrm{spin}}\times\mathrm{SO}(4)_{\mathrm{iso}}$ symmetry. Then, the phase diagram in three spatial dimensions is obtained from a Ginzburg-Landau (GL) free energy whose form is strongly constrained by symmetry,

$$\begin{aligned} F \;&=\; \frac{1}{2}(\nabla P_{\bar{a}\alpha})^2 + \bar{r}P^2_{\bar{a}\alpha} + \delta r\left(P^2_{1\alpha}+P^2_{2\alpha}-P^2_{3\alpha}-P^2_{4\alpha}\right) \\ &+\; \tilde{u}_1 P^2_{\bar{a}\alpha}P^2_{\bar{b}\beta} + \tilde{u}_2 P_{\bar{a}\alpha}P_{\bar{a}\beta}P_{\bar{b}\alpha}P_{\bar{b}\beta} \end{aligned} \qquad (6)$$

This is the most general expression with $\mathrm{SO}(3)_{\mathrm{spin}}\times\mathrm{SO}(4)_{\mathrm{iso}}$ symmetric quartic coefficients. We follow the common assumption that changing the external control parameters of the system, e.g. temperature and pressure, only affects the quadratic coefficients. These are thus allowed to break the symmetry and tune the phase transition. For $\delta r \neq 0$, the symmetry is broken down to $\mathrm{SO}(3)_{\mathrm{spin}}\times\mathrm{SO}(2)_{\mathrm{c}}\times\mathrm{SO}(2)_{\mathrm{t}}$, where $\mathrm{SO}(2)_{\mathrm{c}}$ is generated by the total charge $Q = Q_+ + Q_-$, and $\mathrm{SO}(2)_{\mathrm{t}}$ comes from the lattice translational symmetry[27].

At half-filling, umklapp scattering must be taken into consideration. This is the case for Bechgaard salts, where structural dimerization splits the conduction band into a full lower band, and a half-filled upper band. Umklapp turns two right movers into left movers, and vice versa,

$$\mathcal{H}_3 \;=\; \frac{g_3}{2L}\sum a^\dagger_{+,k+qs}a^\dagger_{+,p-qt}a_{-,pt}a_{-,ks} + h.c. \qquad (7)$$

The term (7) pins the phase of the SDW, reducing it to the real Néel vector $\vec{N} = \mathrm{Re}\vec{\Phi}$ (for $g_3 > 0$). In the presence of umklapp there is still a direct AF/TSC transition at $K_\rho = 1$[1]. On the other hand, Q_+ and Q_- are no longer conserved separately, and $\mathrm{SO}(4)_{\mathrm{iso}}$ symmetry of the free energy is broken. However, to linear order in g_3, $\mathrm{SO}(4)_{\mathrm{iso}}$ is not broken all the way down to $\mathrm{SO}(2)_{\mathrm{c}}$. The contribution of $\mathcal{H}_3$ to the free energy is $\Delta F = \frac{g_3}{L}\left((\mathrm{Re}\vec{\Phi})^2 - (\mathrm{Im}\vec{\Phi})^2\right)$, which preserves a residual symmetry $\mathrm{SO}(3)_{\mathrm{iso}}$, given by the diagonal subgroup of $\mathrm{SO}(4)_{\mathrm{iso}}$ $\approx\mathrm{SO}(3)_R\times\mathrm{SO}(3)_L$[24]. The generators of $\mathrm{SO}(3)_{\mathrm{iso}}$ are $I_a = J^+_a + J^-_a$. Together with $\mathrm{SO}(3)_{\mathrm{spin}}$ invariance, the total symmetry defined by these operators is $\mathrm{SO}(4)\approx\mathrm{SO}(3)_{\mathrm{spin}}\times\mathrm{SO}(3)_{\mathrm{iso}}$.

The justification for considering small g_3 is as follows. The bare value of g_3 is proportional to dimerization, which is only of order of 1% in $(\mathrm{TMTSF})_2\mathrm{PF}_6$[28]. Furthermore, the GL free energy depends on the renormalized value $g_{3,\mathrm{eff}}$ at the crossover scale between 1D and 3D physics, which is even smaller than the bare value of g_3. Umklapp is irrelevant inside the TSC phase, as well as on the AF/TSC phase boundary. Even in the AF phase, where umklapp is a relevant perturbation, g_3 flows near zero before diverging, and this divergence may be cut off by the onset of 3D coupling. Therefore, everywhere close to the AF/TSC phase boundary, we can assume that $g_{3,\mathrm{eff}}$ is small.

SO(4) symmetry unifies AF and TSC orders, which are now combined into a 3×3 tensor order parameter,

$$Q_{a\alpha} \;=\; \begin{pmatrix} (\mathrm{Re}\vec{\Psi})_x & (\mathrm{Im}\vec{\Psi})_x & N_x \\ (\mathrm{Re}\vec{\Psi})_y & (\mathrm{Im}\vec{\Psi})_y & N_y \\ (\mathrm{Re}\vec{\Psi})_z & (\mathrm{Im}\vec{\Psi})_z & N_z \end{pmatrix} \qquad (8)$$

The columns (rows) of $\hat{Q}$ transform as a vector under the spin (isospin) SO(3) algebra, $[S_\alpha, Q_{b\beta}] = i\epsilon^{\alpha\beta\gamma}Q_{b\gamma}$ ($[I_a, Q_{b\beta}] = i\epsilon^{abc}Q_{c\beta}$). In analogy with (6), the GL free energy near the AF/TSC phase boundary is

$$\begin{aligned} F \;&=\; \frac{1}{2}\left(\nabla Q_{a\alpha}\right)^2 + \bar{r}Q^2_{a\alpha} + \delta r(Q^2_{z,\alpha} - Q^2_{x,\alpha} - Q^2_{y,\alpha}) \\ &+\; \tilde{u}_1 Q^2_{a\alpha}Q^2_{b\beta} + \tilde{u}_2 Q_{a\alpha}Q_{a\beta}Q_{b\alpha}Q_{b\beta} \end{aligned} \qquad (9)$$

When $\delta r = 0$ the model has full SO(4) symmetry. Away from this line it only has spin and charge $\mathrm{SO}(3)_{\mathrm{spin}}\times\mathrm{SO}(2)_{\mathrm{c}}$ symmetry. A derivation of the GL free energy for weakly interacting Q1D electrons yields the model in (9) with $\tilde{u}_1 = 21\zeta(3)/16\pi^2 v_f T^2$ and $\tilde{u}_2 = -7\zeta(3)/8\pi^2 v_f T^2$[24].

The properties of model (9) depend strongly on the sign of $\tilde{u}_2$, which determines whether the triplet superconductor is unitary ($\Re\vec{\psi} \propto \Im\vec{\psi}$) or non-unitary ($\Re\vec{\psi}\times\Im\vec{\psi} \neq 0$). We expect the unitary case, $\tilde{u}_2 < 0$, to be of experimental relevance to $(\mathrm{TMTSF})_2\mathrm{PF}_6$, and in the remainder of this paper we primarily concentrate on it. The mean field diagram is then composed of an AF phase separated from a TSC phase by a first order phase transition, and a disordered (Normal) phase separated from the two other phases by second order lines

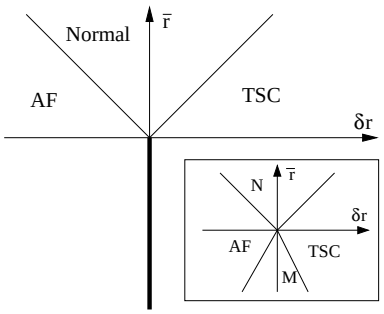

FIG. 1: Mean field phase diagram of eq. (9) in the unitary case $\tilde{u}_2 < 0$. There is a first order transition (thick line) between AF and TSC phases. **Inset:** Corresponding diagram for non-unitary case $\tilde{u}_2 > 0$. M denotes a mixed AF/TSC phase.

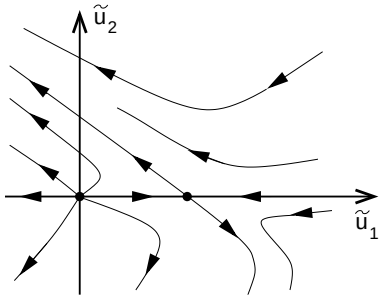

FIG. 2: Renormalization group flow of the SO(4) symmetric theory eq. (9) in $d = 4 - \epsilon$. There are no stable fixed points. Instead, there are two types of runaway flow, corresponding to unitary ($\tilde{u}_2 < 0$) and non-unitary ($\tilde{u}_2 > 0$) TSC.

(see Fig. 1). For completeness, we include in the inset the mean field phase diagram for the non-unitary case.

To understand the role of thermal fluctuations in model (9) and in slightly perturbed models where the quartic coefficients do not lie exactly on the SO(4) symmetric manifold, we use $4 - \epsilon$ RG analysis. We find that the RG equations have only two fixed points: a trivial Gaussian fixed point, $\bar{r} = \delta r = \tilde{u}_i = 0$, and an SO(9) Heisenberg point, $\bar{r} \neq 0$, $\delta r = 0$, $\tilde{u}_1 \neq 0$, $\tilde{u_2} = 0$. In Fig. 2 we show RG flows in the SO(4) symmetric plane, where we find runaway flows whenever $\tilde{u}_2 \neq 0$. The analysis can be generalized to order parameters $\vec{N}$ and $\vec{\Psi}$ that are N-component vectors, in which case the SO(4)≈SO(3)$_{\rm spin}$×SO(3)$_{\rm iso}$ symmetry becomes SO(N)$_{\rm spin}$×SO(3)$_{\rm iso}$. We find that even in the large N limit, all flows with $\tilde{u}_2 < 0$ are runaway flows, indicating the absence of fixed points with unitary TSC.

The absence of a fixed point in the RG flow often implies that fluctuations induce a first order phase transition, thus precluding a multicritical point in the phase diagram. In order to inspect this possibility, we study model (9) directly in 3D in the large N limit[24]. The idea of the large N expansion is to sum self-consistently all bubble diagrams. We find (inset of Fig. 3) a first

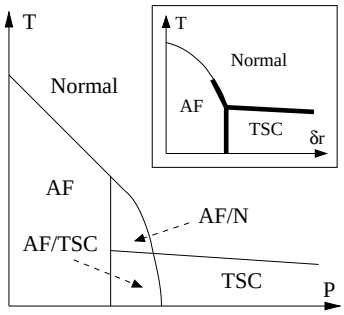

FIG. 3: Schematic temperature-pressure phase diagram of $(TMTSF)_2PF_6$ [12, 13]. AF/N and AF/TSC correspond to coexistence regimes of the appropriate phases. **Inset:** Phase diagram for competing AF and unitary TSC states for model (9) in the large N limit. δr tunes the system across the two phases. Thick lines represent first order transitions.

order transition between AF and TSC phases along the SO(4) symmetric line $\delta r = 0$, in agreement with mean field theory. A new feature of the large N limit is the first order transition between the Normal and AF phases in the vicinity of the critical point. If we assume that the experimentally controlled pressure changes an extensive variable conjugate to δr, such as the volume of the system, the first order transition broadens into a coexistence region of TSC and AF. This is consistent with the experimental phase diagram of $(TMTSF)_2PF_6$ (Fig. 3). An unusual feature of the theoretical phase diagram (inset, Fig. 3) is the first order transition between Normal and TSC phases. This is similar to the fluctuation-driven first order transition between Normal and superfluid phases proposed for ^{3}He by Bailin *et al.*[29]. In bulk ^{3}He, the coherence length is very long and the transition is mean-field like. The fluctuation region is so small that tiny discontinuities at the transition caused by fluctuations would be impossible to observe. In Q1D systems, such as Bechgaard salts, the fluctuation region is expected to be large[30] and the discontinuous nature of the Normal to TSC transition may be experimentally accessible. This transition has been investigated through specific heat measurements in $(TMTSF)_2ClO_4$[31]. The results were interpreted as a mean-field BCS transition, although the amplitude of the specific heat jump was unusually large. Since T_c in these materials is very sensitive to impurities, the extra amplitude in the specific heat jump might be attributed to broadening due to disorder of a δ-function peak in specific heat. Experimental observation of the coexistence region of Normal and TSC phases, or of hysteresis effects in resistivity measurements may also be very difficult. Nearly equal strains in the TSC and the Normal phases and the nearly temperature independent superconducting transition temperature can make such a coexistence region very small. On the other hand, it is possible that spin anisotropy present in real materials, but not included in our theoretical analysis, stops

the runaway RG flows leading to the first order transition. Another possibility is that one loop $4-\epsilon$ RG calculations and the large N expansion do not capture the correct behavior of model (9) for $\epsilon = 1$ and $N = 3$. For the Normal to unitary TSC transition, De Prato *et al.* argued that a stable fixed point describing the second order phase transition appears in the six-loop expansion of the GL free energy[32]. We hope that future experiments will investigate the nature of the Normal to TSC transition in Bechgaard salts in more detail.

The most dramatic consequence of enhanced symmetry at the phase transition is the prediction of new low energy collective excitations. In the vicinity of the AF/TSC phase transition, SO(4) symmetry leads to a new collective mode, corresponding to the Θ operator that rotates AF and TSC orders into each other[24]. As pressure is varied toward the phase transition, breaking of the SO(4) symmetry is reduced, leading to a decrease in the energy of the Θ excitation. Mode softening is not expected generically at a first order phase transition and identifies the Θ-resonance as a generator of the SO(4) quantum symmetry. Weak symmetry breaking due to interchain coupling and higher order umklapp terms may lead to a small gap and to finite broadening of Θ, even at the AF/TSC phase boundary. Deep in the normal phase the Θ excitation cannot be probed by conventional methods, such as electromagnetic waves or neutron scattering, as these only couple to particle-hole channels (e.g. spin or density) and Θ is a collective mode in the particle-particle channel. The situation changes when the system becomes superconducting. In the presence of a condensate of Cooper pairs, charge is not a good quantum number and particle-particle and particle-hole channels mix. The Θ excitation should thus appear as a resonance in inelastic neutron scattering experiments[24], and its intensity should be proportional to the square of the superconducting amplitude $|\vec{\Psi}|^2$. For Q1D Bechgaard salts, we expect strong pairing fluctuations even above T_c. Hence, precursors of the Θ resonance may be visible in the normal state, with strong enhancement of the resonant scattering intensity appearing when long range TSC order develops.

It is useful to put the SO(4) model of AF/TSC competition in Bechgaard salts in the general perspective of electron systems with competing orders. In the case of the SO(5) theory of AF and d-wave SC in 2d systems[7, 9], it is difficult to construct realistic microscopic models with such symmetry (see e.g. Refs. 34, 35). By contrast, SO(4) symmetry in Bechgaard salts arises naturally from a conventional Luttinger description. We also point out that tuning across the AF/TSC phase boundary in $(TMTSF)_2PF_6$ can be done in the same sample by varying pressure, whereas tuning the AF/SC transition in the cuprates requires using different samples. Thus, we consider $(TMTSF)_2PF_6$ a good candidate for experimental observation of emergence of higher symmetry in a strongly correlated electron system.

In the discussion above, we assumed Luttinger liquid behavior in individual chains to motivate the approximate SO(4) symmetry at the AF/TSC phase boundary. It has been suggested that for the superconducting phase of Bechgaard salts, interchain hopping is strong enough to turn the system into a strongly anisotropic Fermi liquid[36, 37]. The decreased nesting condition in this case strongly affects antiferromagnetism. For the classical symmetry of the GL free energy, this effect can be absorbed into the normalization of the field $\vec{N}$, so that the GL parameters only display a small deviation from SO(4) symmetry at the mean-field level. Thus, we do not expect a qualitative change in the phase diagram presented in Fig. 3 (see Ref. [24] for a detailed discussion). To verify the approximate quantum SO(4) symmetry for the strongly anisotropic Fermi liquid, one can study the spectrum of collective excitations using an RPA-type analysis and verify the existence of the Θ excitation[38]. These results will be presented elsewhere.

In summary, we introduced an SO(4) framework for the competition between AF and TSC in Q1D electron systems. The microscopic origin of the SO(4) symmetry at the transition between AF and TSC orders was identified in the Luttinger liquid model. Our results have direct implications for Q1D organic superconductors from the $(TMTSF)_2X$ family. For example, first order transitions between AF and TSC phases, and between AF and Normal phases, explain the AF/TSC and the AF/Normal coexistence regions found in the phase diagram of $(TMTSF)_2PF_6$ [12]. We also argue that the Normal/TSC transition in these materials could be weakly first order. We predict a sharp resonance in neutron scattering, whose characteristics identify it unambiguously as a generator of SO(4) symmetry.

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